BC

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MINERAL RESOURCES DIVISION
Geological Survey Branch

PHOSPHORUS IN BRITISH COLUMBIA COKING COALS

By D.A. Grieve

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ABSTRACT

The concentrations of phosphorus in raw and clean coal samples from the Mist Mountain Formation in the East Kootenay coalfields of southeast British Columbia, and the Gates and Gething formations in the Peace River coalfield in the northeast of the province, have been compiled. Raw coals of the Mist Mountain Formation contain the highest mean phosphorus concentrations (0.076 per cent), Gething Formation coals are intermediate (0.063 per cent), and those of the Gates Formation contain the least (0.042 per cent). Phosphorus is mainly associated with the inorganic fraction of the coals. The principal factors controlling phosphorus contents in raw British Columbia coking coals are the ash content and the types of accessory minerals. Most of the phosphorus is believed to be in apatite minerals, including fluorapatite.

There are no consistent stratigraphic trends in phosphorus contents in raw Mist Mountain coals, although coals from the upper half of the formation appear to contain more phosphorus, on average, than those from the lower half. The basal part of the formation hosts coals with relatively low phosphorus (and apatite) contents. In the case of the Gates Formation, coal seams from the upper part of the section contain, on average, higher phosphorus concentrations than those from the lower part. In all three coal-bearing formations, the amount of data scatter within individual seams can be very pronounced.

The range in average clean coal phosphorus concentrations, 0.030 to 0.044 per cent, is smaller than the range in average raw concentrations for the three formations. Clean Mist Mountain and Gething Formation coals contain substantially less phosphorus than corresponding raw coals, while the mean in clean Gates coals is essentially unchanged from the value in raw coals. This contrast is at least partly related to analytical procedures, and does not reflect fundamental differences in the quality characteristics of Gates Formation coals. The two coal mining regions, northeast and southeast British Columbia, ship coking coals with very similar phosphorus concentrations. A representative average of phosphorus concentrations in clean British Columbia coking coals, including products from currently producing mines and results of this study, is 0.05 per cent.

EAST KOOTENAY COALFIELDS
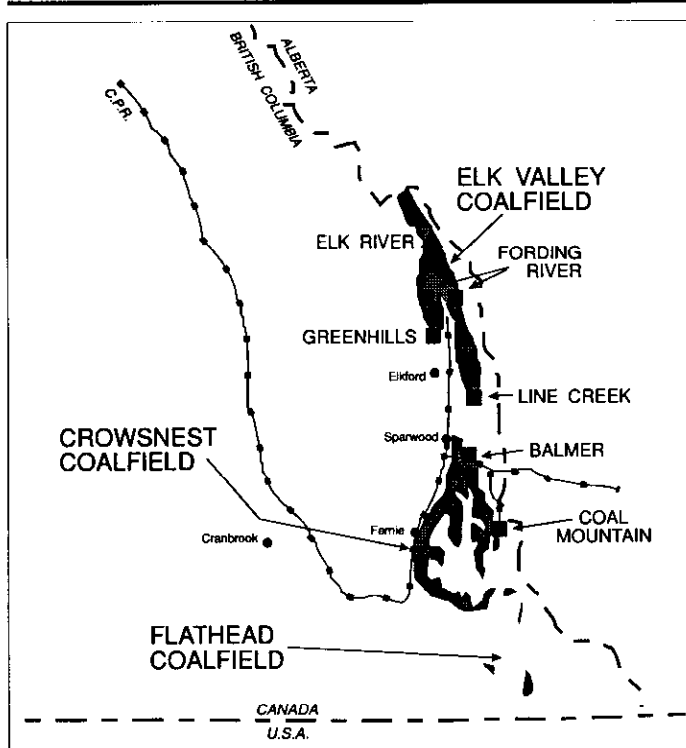
PEACE RIVER COALFIELD

LEGEND

●—●—● Railways
● Communities
■ Mines
▲ Other coal
   deposits in study

LEGEND

●—●—● Railways
● Communities
■ Mines
▲ Other coal
   deposits in study

ALBERTA
BRITISH COLUMBIA
C.P.R.

ELK VALLEY
COALFIELD

ELK RIVER

FORDING
RIVER

GREENHILLS

Elkford

LINE CREEK

CROWSNEST
COALFIELD

Sparwood

BALMER

Cranbrook

Fernie

COAL
MOUNTAIN

FLATHEAD
COALFIELD

CANADA
U.S.A.

E. & S. MT GETHING

CARBON
CREEK

B.C.R.

Chetwynd

Dawson Creek

SUKUNKA

GOODRICH

MT SPIEKER

Tumbler Ridge

BULLMOOSE

QUINTETTE

Anzac

MONKMAN

BELCOURT

B.C.R.

C.N.R.

SAXON

Prince
George

BOWRON RIVER
COALFIELD

ALBERTA
BRITISH COLUMBIA
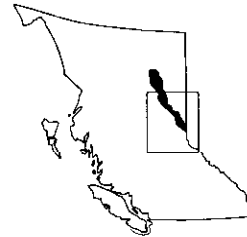
Figure 1. Locations of coking coal mines and deposits referred to in this study.

INTRODUCTION

Phosphorus occurs in all coals in minor or trace amounts. Although it is not generally regarded as a pollutant, the concentration of phosphorus in a coal is an important parameter to coal users, particularly steel mills. Phosphorus in steel, some of which is derived from coke, can be either beneficial or detrimental to its quality. Small additions of phosphorus are sometimes used to increase the strength of low-carbon sheet steel (Bloom *et al.*, 1990). However, under certain conditions, phosphorus addition will cause steel to become embrittled. There is no universally accepted tolerance level for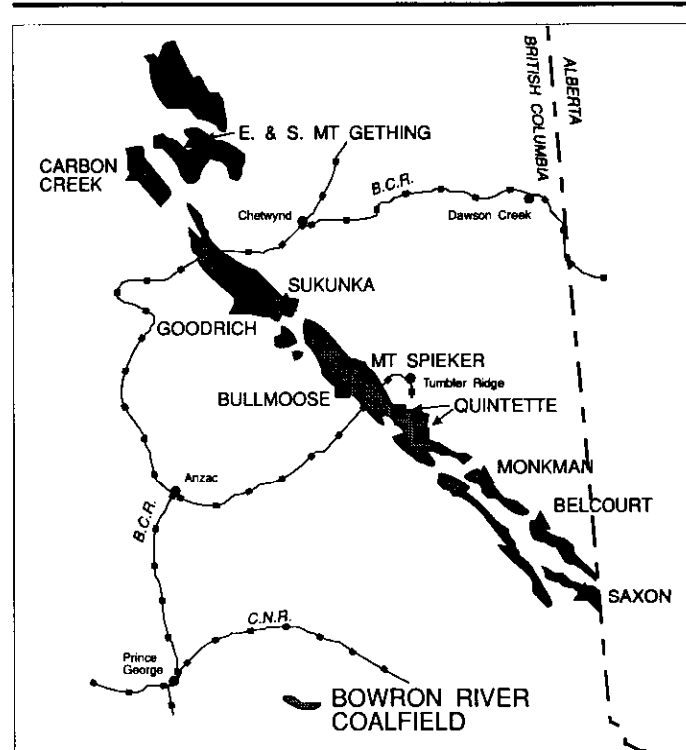 phosphorus in coking coal, and in some cases it is not a major concern. Other variables are equally or more critical, including the phosphorus concentrations in the iron ore and the steel-making process used. Iron formations of sedimentary origin, for example, contain much less phosphorus than iron ores of igneous origin. Phosphorus content is also of interest to operators of some coal-fired boilers, because, under certain conditions, phosphate precipitates may form (Burchill *et al.*, 1990).

To date in this study, phosphorus contents of British Columbia coking coals have been compiled for the purposes of comparison and making preliminary conclusions concerning form(s) of association of phosphorus. Follow-up work will investigate the association of phosphorus in more detail.

Coking coal in British Columbia occurs in two regions, the East Kootenay or southeast, and the Peace River or northeast (Figure 1). In the Kootenays, economic coals belong to the Mist Mountain Formation of the Jurassic-Cretaceous Kootenay Group. There are currently four mines in the East Kootenay coalfields which produce coking coal: the Fording River, Balmer, Greenhills and Line Creek mines. A fifth mine, Coal Mountain, produces and markets weak coking coal. In the Peace River coalfield, major bituminous coal resources belong to the Gething Formation of the Lower Cretaceous Bullhead Group, and the younger Gates Formation of the Lower Cretaceous Fort St. John Group. There are two mines in the Peace River area, both of which produce coking coal: the Quintette and Bullmoose mines. In both cases the coal is hosted by the Gates Formation.

DATA COLLECTION

Data used in this paper come from two main sources, assessment reports, written by industry geologists to document exploration results on individual coal properties and deposits, and analysis of a set of channel samples collected by the author at all seven of the mines in the Peace River and East Kootenay coalfields (Figure 1) in 1989. The assessment report data are the more representative, because they are based on many more readings, and include the only Gething Formation and clean coal data available. The author's samples provide an opportunity to compare concentrations of phosphorus with those of other trace elements and with the mineralogy of low-temperature ash.

ASSESSMENT REPORT DATA

Exploration assessment reports were used to compile databases of phosphorus concentrations in raw and clean coals. In some cases data on both raw and clean fractions of the same sample are available. The exploration properties represented by these data are listed in Table 1 and their locations are shown on Figure 1. Ash

and sulphur data were also collected for each record. Most of the data represent analyses of drill-core samples. In most cases, data were reported as per cent phosphorus in coal, although for some properties the P values in coal had to be calculated from the concentration of P_2O_5 in coal ash. Most data were available on an air-dried basis, and those that were not were converted. Data in reports for which the basis of reporting was not explicitly defined were not included.

SAMPLING

Channel samples were collected from fresh coal faces in active mines (*see* Figure 1 and Table 1) for trace element analyses, and to provide samples for low-temperature ashing (Grieve and Holuszko, 1991). The channels were approximately 10 centimetres wide by 5 centimetres deep. Each sample was intended to be representative of a seam or interval. In the cases of six of the seven mines, whole-seam samples were collected, for a total of thirty samples, twenty-two from southeast British Columbia and eight from the northeast. At the other mine, Line

TABLE 1
COAL DEPOSITS COVERED IN THIS STUDY

REGION	FORMATION	PROPERTY/ DEPOSIT NAME	SAMPLE TYPE	No. of SAMPLES	SOURCE OF DATA
Southeast	Mist Mountain	Elk River	raw & clean	51	Assessment report 274
		Elk River	clean	194	Assessment report 276
		Fording River	raw, whole-seam	7	channel sampling program
		Greenhills	raw, whole-seam	6	channel sampling program
		Line Creek, 10A-seam	raw, ply-by-ply	5	channel sampling program
		Line Creek, 10B-seam	raw, ply-by-ply	5	channel sampling program
		Line Creek, 9-seam	raw, ply-by-ply	8	channel sampling program
		Line Creek, 8-seam	raw, ply-by-ply	19	channel sampling program
		Balmer	raw, whole-seam	6	channel sampling program
		Coal Mountain	raw, whole-seam	3	channel sampling program
Northeast	Gething	Carbon Creek & Carbon Creek W.	raw	56	Assessment reports: 504, 507 & 508
		E. Mt. Gething & S. Mt. Gething	raw	11	Assessment reports: 520 & 639
		Goodrich	raw	21	Assessment reports: 532 & 534
		Sukunka	clean	48	Assessment reports: 645 & 650
		Mt. Spieker	clean	15	Assessment reports: 552, 553 & 555
Northeast	Gates	Bullmoose	raw, whole-seam	6	channel sampling program
		Mt. Spieker	clean	36	Assessment reports: 552, 553, 555, 556 & 558
		Quintette	raw & clean	303	Assessment reports: 603, 611, 615 & 619
		Quintette	raw, whole-seam	2	channel sampling program
		Monkman	clean	43	Assessment reports: 543, 545, 546 & 547
		Belcourt	raw & clean	97	Assessment reports: 463 & 466
		Saxon	raw & clean	86	Assessment report 628

Creek, four major seams, 10A, 10B, 9 and 8, were sampled in plies averaging 50 cemtimetres in thickness, for a total of 37 samples.

SAMPLE PREPARATION AND ANALYSIS

Samples were dried, crushed, riffled and screened in accordance with ASTM standard procedures. Proximate and other routine analyses were also carried out using ASTM standardized techniques. Phosphorus in coal was determined by ASTM method D2795, and reported as per cent P_2O_5. These values were converted to per cent phosphorus in coal. Fluorine in coal was determined by ASTM method D3761.

Representative splits of -60-mesh coal were provided to the author for low-temperature ashing. This technique, which uses radio frequency (RF) generated oxygen plasma, is a routine way of producing an ash with the original minerals essentially preserved (*see* for example, Miller *et al.*, 1979). The Ministry's plasma asher is an LFE Corporation model LTA-504, which uses an RF power supply that operates at 13.56 megahertz. Five to ten grams of coal are placed in a silica sample-boat. One boat is placed in each of the four 10-centimetre diameter reaction chambers, which are then evacuated using a vacuum pump. Ashing is done using 200 watts total RF power (50 watts per chamber), and a total oxygen bleed-rate of about 30 cubic centimetres per minute. Samples are left exposed to the oxygen plasma round-the-clock, for a total of about 72 hours. They are stirred at least twice a day using a glass rod, in order to bring unreacted coal to the surface. At the end of the reaction time a small amount, less than 1 per cent (estimated) by volume, of unreacted organic material is left in the residue. This is assumed to be made up of inertinite. Low-temperature ashes are ground using an agate mortar and pestle, prior to x-ray diffraction analysis.

DETERMINATION OF ORGANIC/ INORGANIC ASSOCIATION IN COAL

The mode of association of a trace element in coal is as important as its actual concentration (Finkelman,

1980). One generally accepted and simple method of gaining a preliminary impression of this factor is to plot the concentration of the element versus the ash content (Nicholls, 1968). If the concentration increases with increasing ash, it can be tentatively concluded that the element is predominantly associated with the inorganic fraction of the coal. Conversely, a constant or decreasing concentration of the element suggests an association with the organic fraction. A further refinement is to divide the concentration of the element in each sample by the ash content of the sample, in effect calculating the element's concentration in ash, and to plot this value against the ash. With this approach, element concentrations which plot on a horizontal line suggest a predominantly inorganic association, while those which plot on a line with negative slope imply organic association. Nicholls (1968) also makes a case for being able to detect a mixed association with this latter type of graph. Figure 8f is an example of the type of plot which has been interpreted in this way. It shows a high negative slope at low ash contents (supposedly organic association), and no slope over higher ash contents (inorganic). Some of the assumptions inherent in the use of element/ash graphs are suspect, particularly the assumption that the element's concentration is consistent in each ash increment (Nicholls, 1968). These assumptions will be evaluated later, with examples from this study.

A second widely used approach for determining an element's affinity in coal is to compare its concentration in specific gravity fractions from sink-and-float testing (Gluskoter *et al.*, 1977). Elements with organic affinity should be concentrated in the light fraction(s), while those with inorganic affinity should be enhanced in the heavy fraction(s). This assumes, however, that mineral grains are physically liberated prior to the separation. Problems with this assumption will also be discussed later.

Other methods, such as SEM-EDX analysis (Van der Flier-Keller and Fyfe, 1987), have not been attempted to date. Future work will utilize some of the other available techniques to determine affinities of phosphorus and other trace elements.

RESULTS

Data are represented in both tabular (Tables 2 to 4) and graphic form (Figures 2 to 11). Complete analytical results on the channel samples collected in 1989 are included as Appendix 1.

PHOSPHORUS CONCENTRATIONS IN RAW COALS

MIST MOUNTAIN FORMATION

Raw Mist Mountain coals (from the East Kootenay coalfields of southeast British Columbia) have mean phosphorus concentrations of 0.076 per cent in the case of assessment report data, and 0.096 per cent in the case of the whole-seam channel samples.

Variations in phosphorus concentration with stratigraphic position in the Mist Mountain Formation are shown in Table 2 and Figures 2a and 3. An up-section increase in phosphorus contents has been noted at one location (B. Ryan, personal communication, 1988). However, none of the data shown here suggest that there is any systematic variation in phosphorus values. General stratigraphic trends are evident, however. For example, data in Figure 2a (channel samples) suggest that phosphorus

contents are lowest near the base and top of the formation, and highest in the middle. This pattern, however, is almost identical to that of the ash contents of the same samples (Figure 2b), suggesting that ash content is perhaps a more important determinant of phosphorus content than stratigraphic position. The assessment report data suggest a somewhat different pattern (Table 2 and Figure 3). With the exception of 3-seam, the seams with the greatest phosphorus concentration tend to be in the upper half of the formation. The basal seam (number 2) on the Elk River property has the lowest phosphorus contents, which is consistent with data in Figure 2a. One obvious conclusion to be derived from Table 2 and Figure 3 is that there is a great deal of variation in raw phosphorus concentrations between samples of the same seam.

In the cases of the four seams at Line Creek, numbers 9 and 8 have higher average phosphorus contents than the underlying 10A and 10B-seams at the base of the formation, but there are no consistent in-seam variations (Figures 2c and 2d).

GETHING FORMATION

Raw Gething Formation coals (from the Peace River coalfield) have a mean phosphorus concentration (air dried) of 0.063 per cent; all data are from assessment reports. No stratigraphic trends in phosphorus contents are apparent.

GATES FORMATION

Raw Gates Formation coals (from the northeast) have mean phosphorus concentrations of 0.042 per cent in the case of assessment report data, and an almost identical 0.043 per cent in the case of the whole-seam channel samples. Assessment report data suggest that mean raw coal phosphorus concentrations tend to be higher in seams in the upper part of the Gates Formation than in the lower (Table 3 and Figure 4). Again, the amount of variation of phosphorus values within data for some individual seams is very striking. This upward trend is basically confirmed by the channel sample data (Figure 2e). As was the case with the Mist Mountain channel samples, however, the stratigraphic profile of phosphorus concentrations in channel samples matches the profile of ash contents in the same samples fairly closely (Figure 2f).

TABLE 2
STRATIGRAPHIC VARIATIONS IN PHOSPHORUS
CONCENTRATIONS IN RAW MIST MOUNTAIN
FORMATION COALS*
(ELK RIVER PROPERTY)

SEAM	No. of SAMPLES	MEAN P (%)	RANGE IN P (%)
2	2	0.021	0.020 - 0.022
3	3	0.114	0.049 - 0.183
4	4	0.027	0.015 - 0.039
6	1	0.090	-
7	2	0.062	0.057 - 0.066
8	2	0.072	0.056 - 0.088
9	2	0.030	0.028 - 0.031
10	2	0.059	0.045 - 0.073
12	3	0.087	0.072 - 0.109
13	10	0.096	0.047 - 0.152
14	6	0.059	0.039 - 0.078
15	2	0.088	0.079 - 0.096
16	6	0.105	0.056 - 0.151
17	3	0.094	0.084 - 0.113
18	2	0.074	0.073 - 0.075

*Seams listed in ascending stratigraphic order;
all data from assessment reports.*
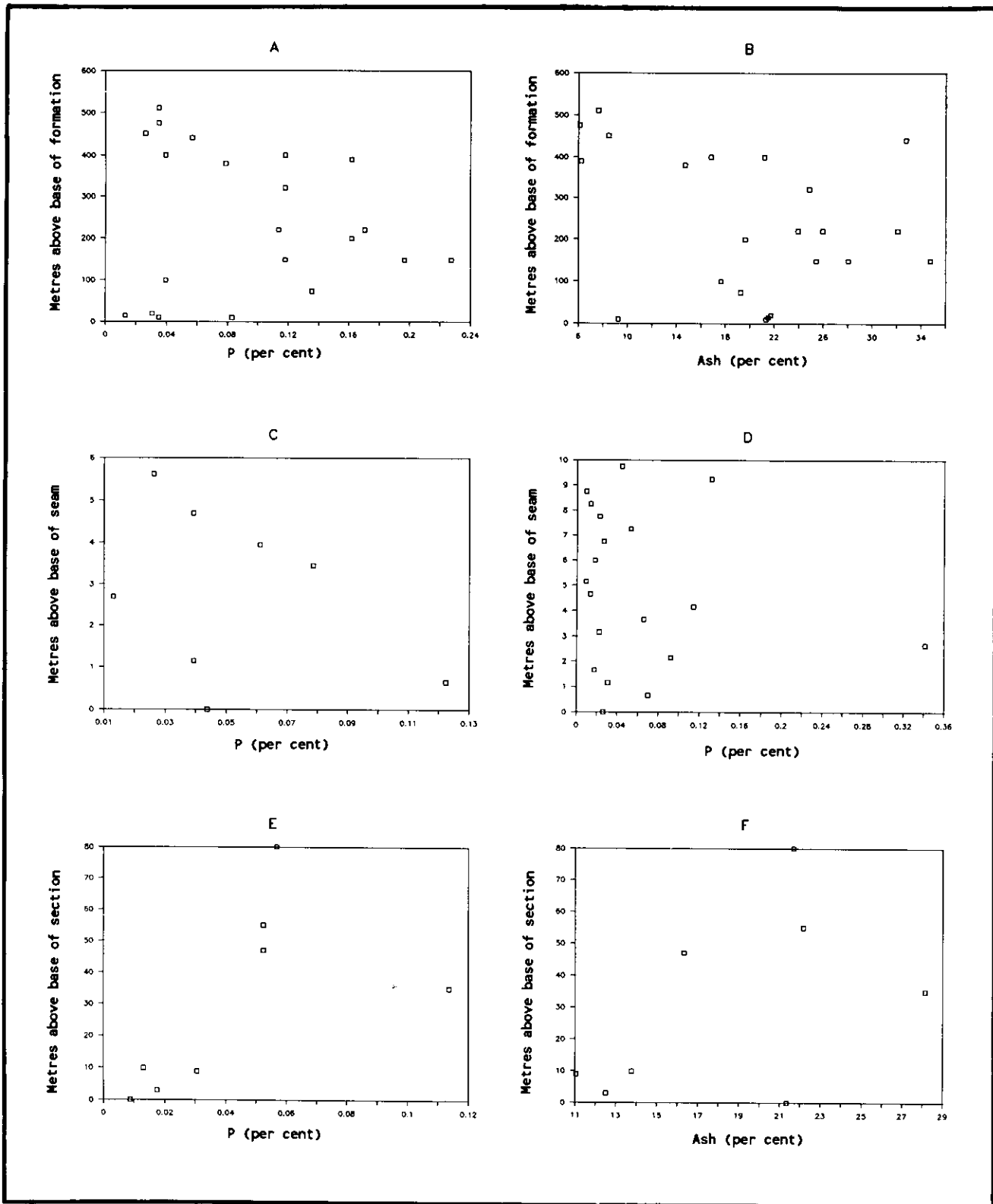
Figure 2. Variations in phosphorus concentrations and ash contents with approximate stratigraphic position, for raw, channel samples collected from active mines. (A): phosphorus, East Kootenay coalfields; (B): ash, East Kootenay coalfields; (C): phosphorus, Line Creek 9-seam; (D): phosphorus, Line Creek 8-seam; (E): phosphorus, Peace River coalfield; (F): ash, Peace River coalfield.
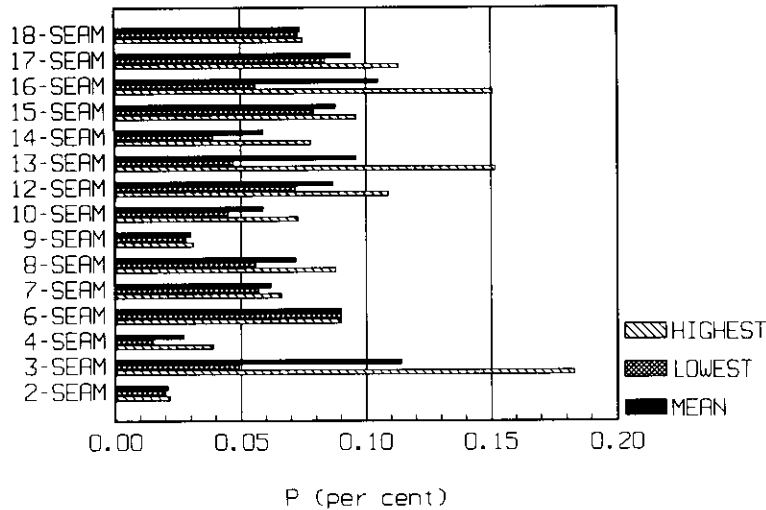
Figure 3. Means, highest and lowest phosphorus concentrations in coal seams of the Mist Mountain Formation (Elk River property). Seams are arranged in stratigraphic order.

TABLE 3
STRATIGRAPHIC VARIATIONS IN PHOSPHORUS
CONCENTRATIONS IN RAW GATES FORMATION
COALS*

PROPERTY	SEAM	No. of SAMPLES	MEAN P (%)	RANGE IN P (%)
Belcourt	1	18	0.026	0.009 - 0.051
	2	14	0.026	0.011 - 0.084
	3	4	0.029	0.018 - 0.052
	4	5	0.014	0.008 - 0.022
	5	11	0.025	0.007 - 0.047
	6	20	0.063	0.004 - 0.224
	7	9	0.042	0.002 - 0.145
	8	16	0.052	0.020 - 0.124
Saxon	1	27	0.014	0.002 - 0.044
	2	18	0.019	0.006 - 0.035
	3	10	0.025	0.006 - 0.079
	4	25	0.021	0.005 - 0.049
	5	3	0.020	0.010 - 0.033
	10	2	0.043	0.031 - 0.054

*Seams listed in ascending stratigraphic order; all data from assessment reports.

STATISTICAL RELATIONSHIPS BETWEEN PHORPHORUS AND ASH

Correlation coefficients (r) between phosphorus and ash contents in raw British Columbia coking coals are listed in Table 4. Graphs illustrating some examples of these relationships, together with corresponding relationships between phosphorus in ash *versus* ash, are shown in Figures 5 to 9. In the case of the assessment report data, r-values are low and there are very few instances where there is significant correlation between phosphorus and ash content (Table 4). F and K-seams at Quintette, however, do show a significant positive correlation at the 99 per cent confidence level (Figure 9a), while the Gates Formation as a whole (Figure 7a), and the Gething Formation (Figure 6a), show a significant relationship at the 95 per cent level. In the case of the channel

sample data, both the Gates and Mist Mountain formations show a significant correlation at the 95 per cent confidence level (Table 4; Figures 5c and 7c). This is consistent with the similarity between the stratigraphic profiles of phosphorus and ash in the channel sample data (Figure 2) noted above.

Consistent with the general lack of strong correlations, interpretation of the plots of phosphorus *versus* ash (Figures 5 to 9) is not straightforward. There is a large amount of data scatter in nearly all cases, and few examples which allow application of Nicholl's (1968) general principles of element affinity, outlined earlier. Some of the graphs of phosphorus in ash *versus* ash, however, appear to be similar to the graphs which supposedly demonstrate both organic and inorganic association. For examples, *see* Line Creek 8-seam (Figure 5h), Gething Formation in total (6b), Gates Formation in total (Figure 7b), Quintette, Belcourt and Saxon properties (Figures 8b, d and f), Quintette F-seam (9b), Belcourt 8-seam (9d) and Saxon 1-seam (9f). These graphs all display a high negative slope at low ash concentrations (the "organic" component), and a zero slope at higher ash levels ("inorganic"). Comments concerning the dubious validity of this interpretation are included under "Discussion."

LOW-TEMPERATURE ASH MINERALOGY OF RAW COALS

The suites of minerals in low-temperature ash derived from the channel samples are listed in Appendix 1. Quartz, kaolinite, illite and carbonates, in order of decreasing abundance, are generally the most common minerals in raw British Columbia coking coals. Of interest to this study is the identification of accessory apatite in many of the samples. Samples in which apatite was positively identified in greater than trace amounts in Line Creek 9 and 8-seams are highlighted in Figures 3f and 3h. Apatite
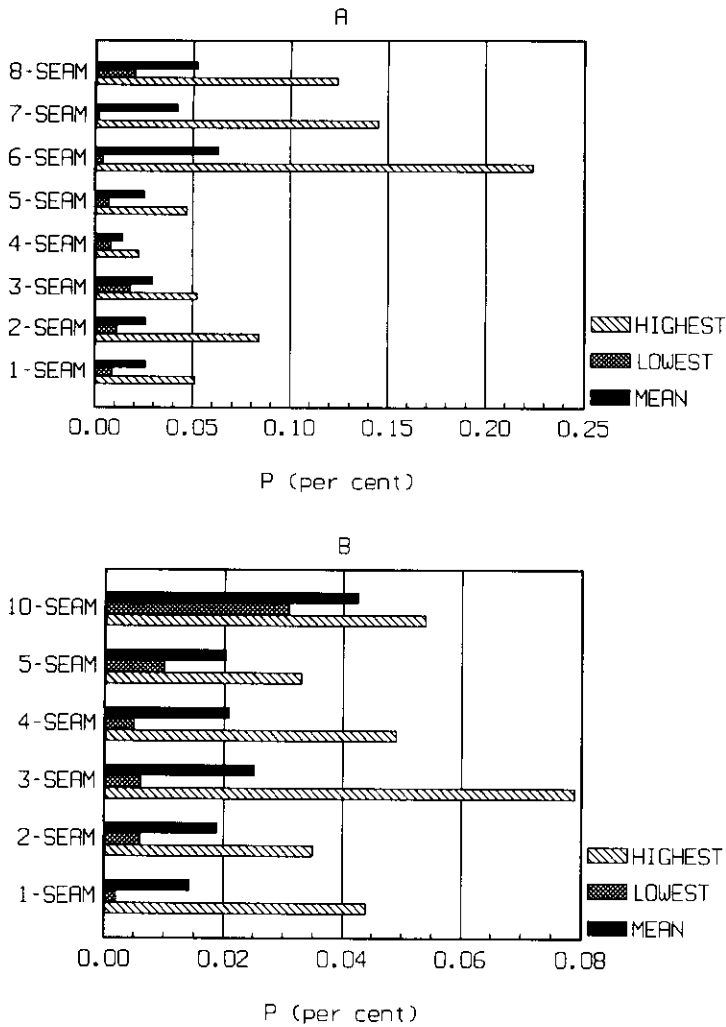
Figure 4. Means, highest and lowest phosphorus concentrations in coal seams of the Gates Formation. Seams are arranged in stratigraphic order. (A): Belcourt property; (B): Saxon property.

TABLE 4
CORRELATION COEFFICIENTS AND LEVELS OF SIGNIFICANCE BETWEEN
PHOSPHORUS AND ASH IN RAW COALS

| REGION/ FORMATION	PROPERTIES/DEPOSITS REPRESENTED	DATA SOURCE	r	95% CONFIDENCE	99% CONFIDENCE	No. of SAMPLES
Southeast	Elk River	Assessment report	0.205	no	no	51
Southeast	Fording, Greenhills, Balmer, Coal Mtn.	Channel samples	0.412	yes	no	22
Southeast	Line Creek 8-seam ply-by-ply	Channel samples	-0.268	no	no	19
Northeast/Gething	Carbon Creek, Mt. Gething, Goodrich	Assessment reports	0.239	yes	no	88
Northeast/Gates	Quintette, Belcourt, Saxon	Assessment reports	0.104	yes	no	486
Northeast/Gates	Quintette	Assessment reports	0.109	no	no	303
Northeast/Gates	Quintette K-seam	Assessment reports	0.666	yes	yes	32
Northeast/Gates	Quintette J-seam	Assessment reports	0.116	no	no	69
Northeast/Gates	Quintette I-seam	Assessment reports	-0.138	no	no	25
Northeast/Gates	Quintette G-seam	Assessment reports	0.117	no	no	31
Northeast/Gates	Quintette F-seam	Assessment reports	0.463	yes	yes	52
Northeast/Gates	Quintette E-seam	Assessment reports	-0.233	no	no	49
Northeast/Gates	Quintette D-seam	Assessment reports	0.107	no	no	40
Northeast/Gates	Belcourt	Assessment reports	-0.024	no	no	97
Northeast/Gates	Saxon	Assessment reports	0.149	no	no	86
Northeast/Gates	Quintette and Bullmoose	Channel samples	0.730	yes	no	8
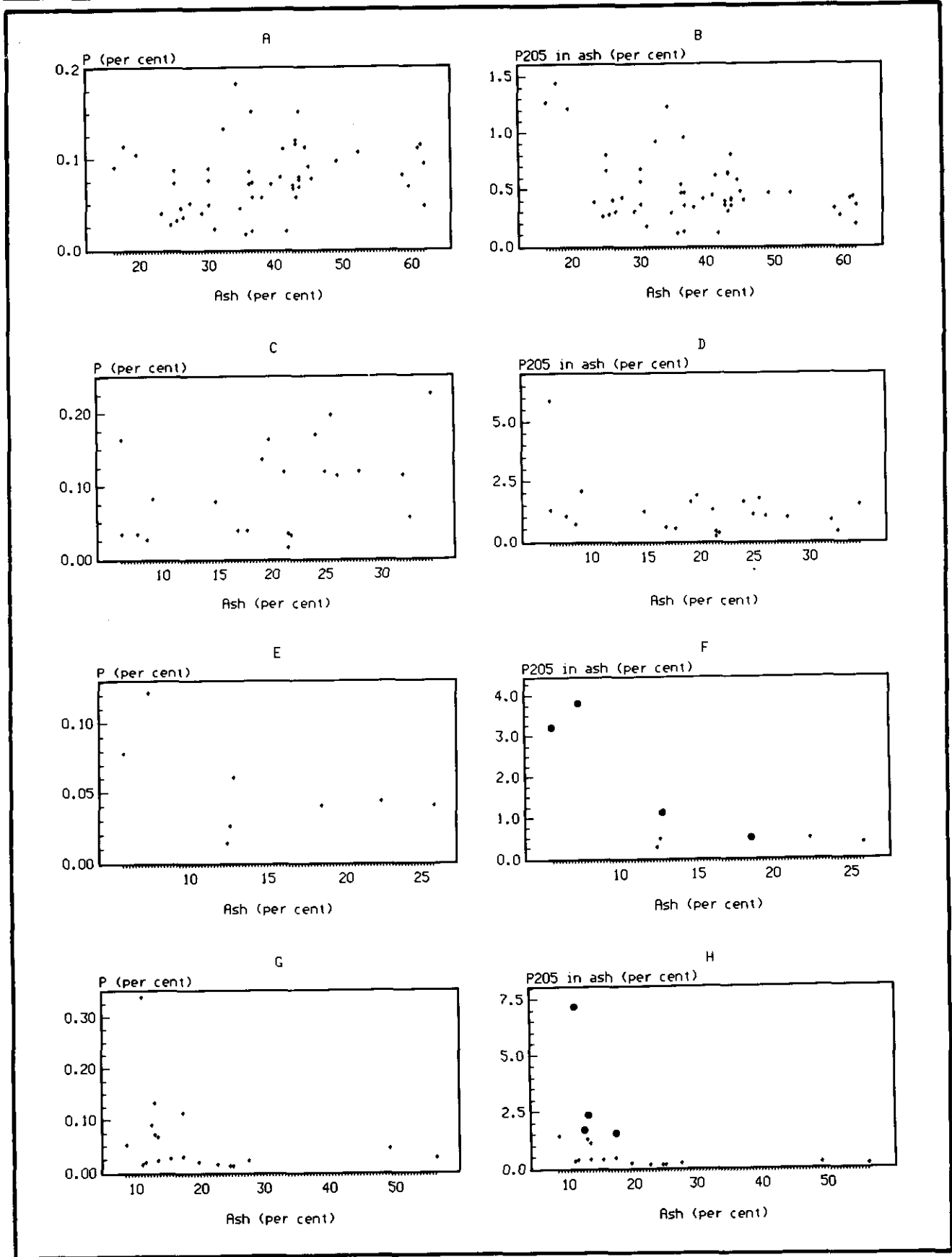
Figure 5. Variations in phosphorus concentration with ash content, raw coals, Mist Mountain Formation, East Kootenay coalfields. (A) and (B): assessment report data; (C) and (D): whole-seam channel samples; (E) and (F): Line Creek 9-seam ply-by-ply channel samples; (G) and (H): Line Creek 8-seam ply-by-ply channel samples. Large dots in (F) and (H) represent samples in which fluorapatite was positively identified in greater than trace amounts in low-temperature ash.

Figure 6. Variations in phosphorus concentration with ash content, raw coals, Gething Formation, Peace River coalfield. All data are from assessment reports.

Figure 7. Variations in phosphorus concentration with ash content, raw coals, Gates Formation, Peace River coalfield. (A) and (B): assessment report data; (C) and (D): whole-seam channel samples.

Figure 8. Variations in phosphorus concentration with ash content, raw coals, Gates Formation, Peace River coalfield. All data are from assessment reports, and represent subsets of data in Figure 7A and 7B. (A) and (B): Quintette; (C) and (D): Belcourt; (E) and (F): Saxon.
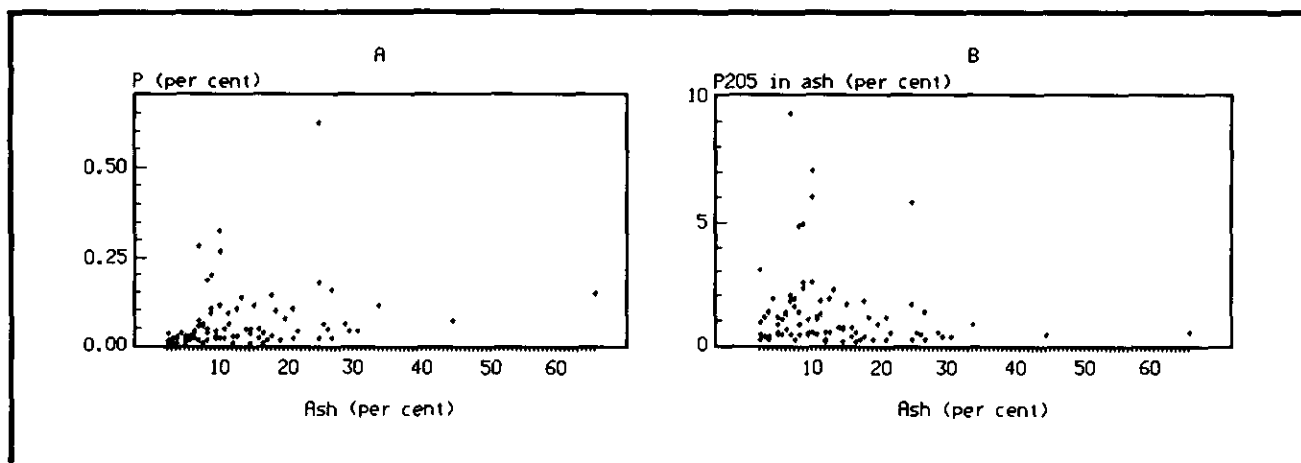
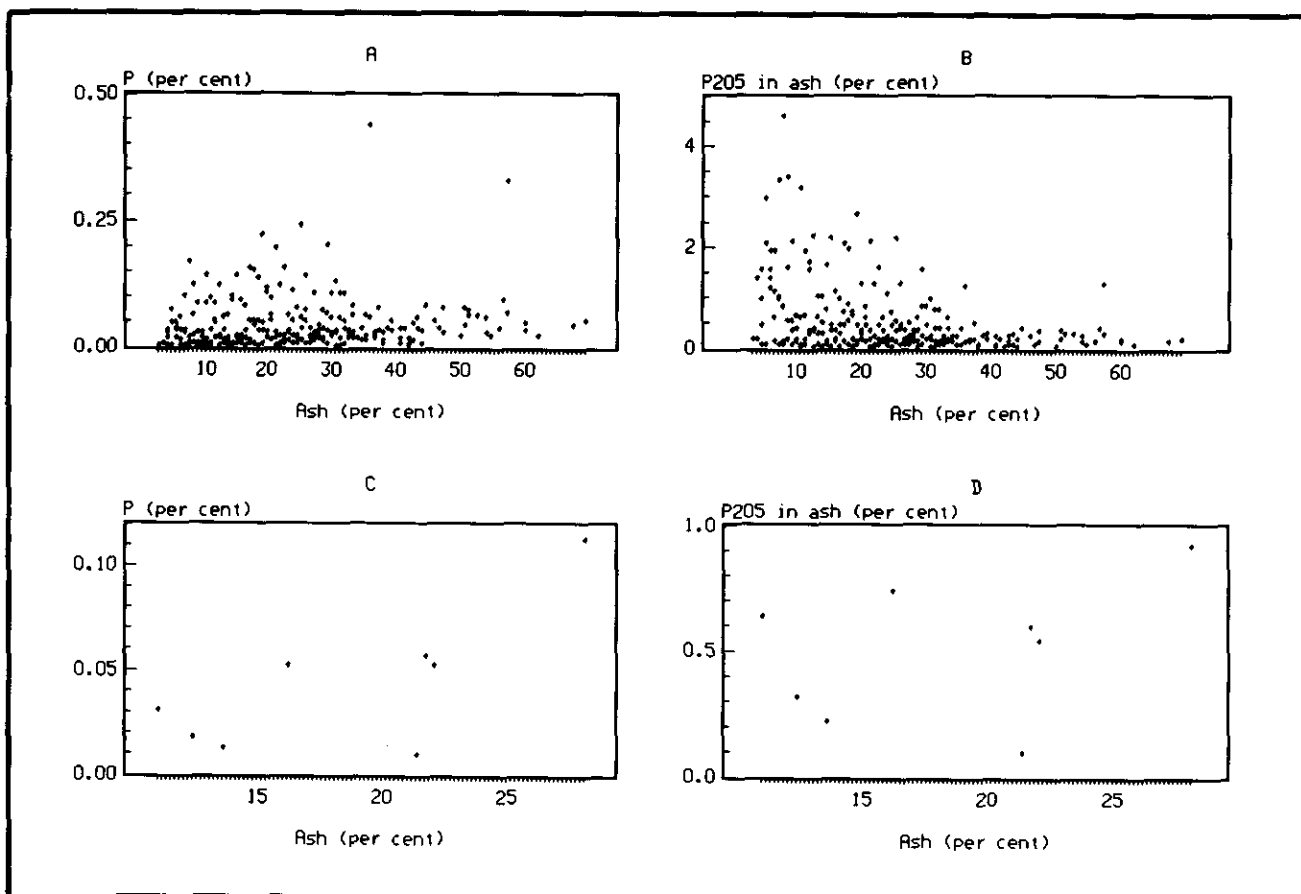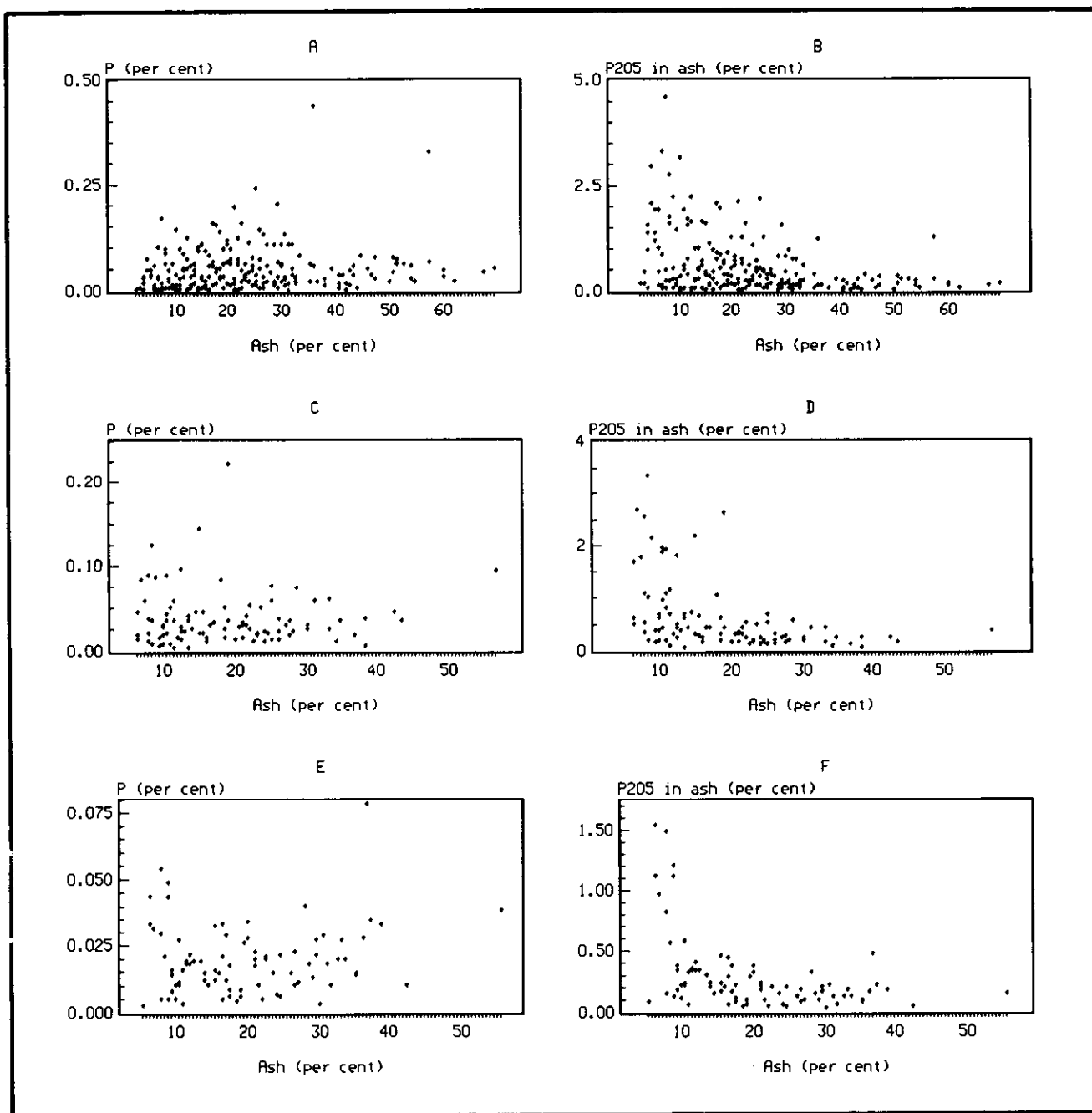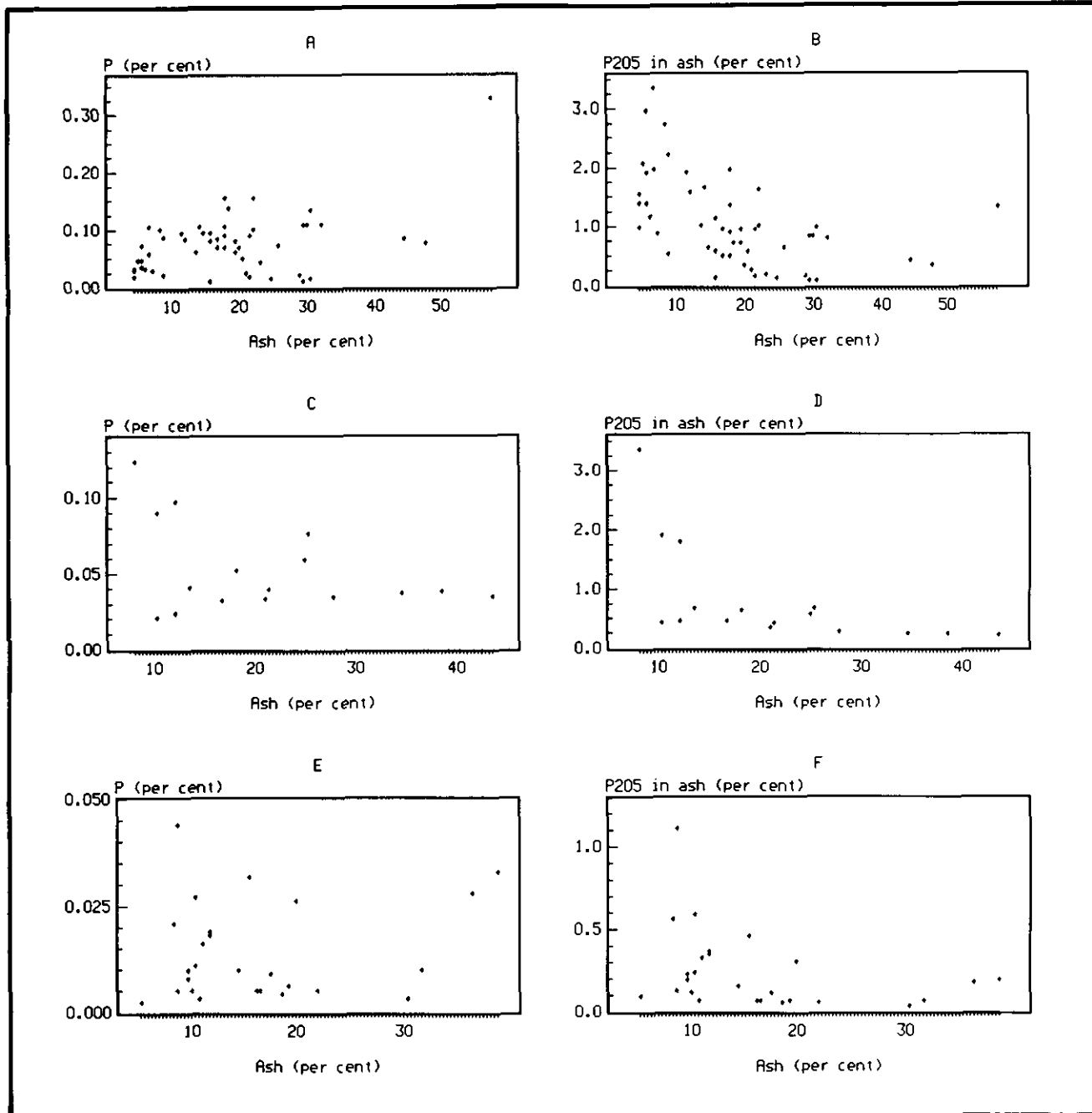Figure 9. Variations in phosphorus concentration with ash content, raw coals, Gates Formation, Peace River coalfield. All data are from assessment reports, and represent subsets of data in Figure 7A and 7B. (A) and (B): Quintette F-seam; (C) and (D): Belcourt 8-seam; (E) and (F): Saxon 1-seam.

minerals appear to be more common in Mist Mountain Formation coal samples than in Gates Formation coals; in fact, trace apatite was only tentatively identified in two out of eight Gates Formation samples. The fluorapatite variety of apatite was frequently identified in Mist Mountain coals, but not in the Gates coals. Samples from seams in the lowermost part of the Mist Mountain Formation, in particular 10A and 10B-seams from Line Creek and the Mammoth seam from Coal Mountain, do not contain detectable apatite. The phosphate mineral gorceixite, a member of the crandallite series, was tentatively identified in the low-temperature ash of one Fording River sample.

RELATIONSHIPS BETWEEN PHOSPHORUS AND FLUORINE

The concentration of other trace elements, including fluorine, in the channel samples was also determined (Grieve and Goodarzi, *in preparation*). A positive correlation between phosphorus and fluorine would be expected if a significant fraction of the phosphorus occurs in fluorapatite. Figure 10 displays phosphorus versus flu-

orine plots; the two elements are positively correlated at the 99 per cent confidence level in all examples shown, except for the Gates Formation samples (Figure 10d), for which the confidence level is 95 per cent. The correlation coefficients are as follows: Mist Mountain Formation (Figure 8a), 0.92; Line Creek 9-seam (8b), 0.88; Line Creek 8-seam (8c), 0.76; Gates Formation (8d), 0.72.

PHOSPHORUS CONCENTRATIONS IN CLEAN COALS

For the Mist Mountain Formation the mean phosphorus concentration in clean coals, based on assessment report data, is 0.033 per cent, while the mean phosphorus concentration in clean Gething Formation coals is 0.030, and in clean Gates Formation coals the value is 0.044 per cent. In the last case, the value is essentially unchanged from the raw coal value, while for the other two the concentration in clean coals represents a significantly lower value than that in raw coals. This comparison is misleading, however, because of differences in analytical procedures between different exploration companies. Most of the Gates samples were crushed to a coarser
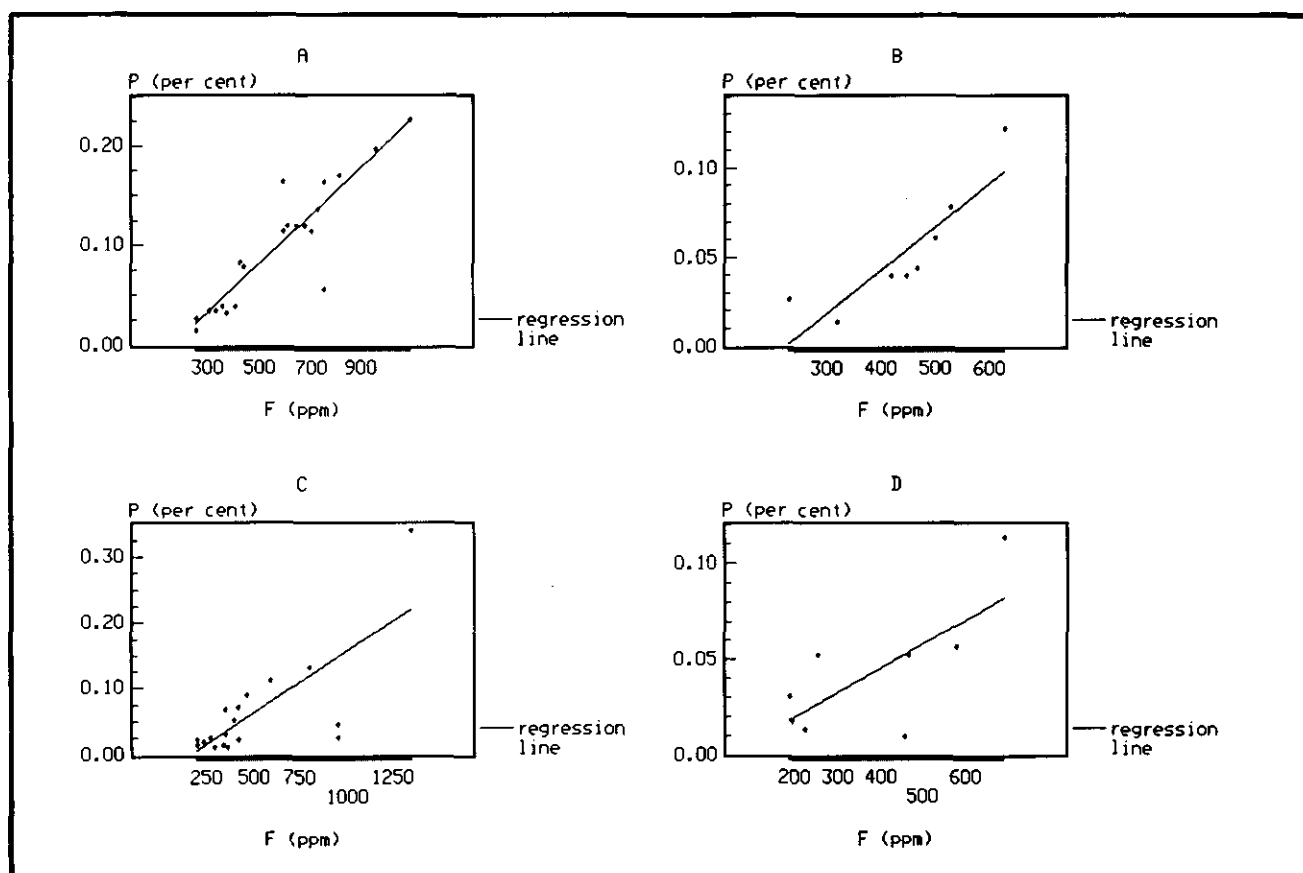

Figure 10. Variations in phosphorus with fluorine concentrations, based on raw channel samples. (A): East Kootenay coalfields, whole-seam samples (Mist Mountain Formation); (B): Line Creek 9-seam, ply-by-ply samples (Mist Mountain Formation); (C): Line Creek 8-seam, ply-by-ply samples; (D): Peace River coalfield, whole-seam samples (Gates Formation).

INCREASE
NO CHANGE
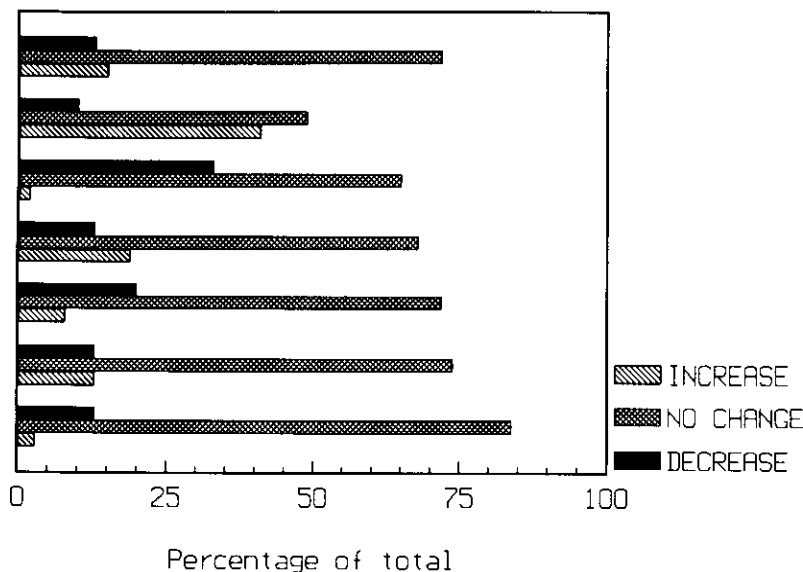DECREASE

Percentage of total

Figure 11. Changes in phosphorus concentrations from raw to clean coal for a Gates Formation deposit. Seams are arranged in stratigraphic order.

maximum size and were floated in higher specific gravity media than the Mist Mountain coals. This would tend to hinder liberation and separation of mineral matter in the Gates samples.

In certain cases, clean and raw phosphorus values are available for the same set of samples. Where this occurs a new variable, delta P, was derived for each sample, by subtracting the raw phosphorus concentration from the clean value. Delta P values between -0.01 and +0.01 per cent are interpreted to represent no change, as the error in each reading is assumed to be 0.005 per cent. For the Mist Mountain Formation, phosphorus concentration decreased (that is, delta P < -0.01) when the coal was washed in 39 out of 51 samples, and increased in only two cases. For the Gates Formation the situation is somewhat different, with the majority of samples, 285 out of a total of 486, experiencing no change, 79 samples showing a decrease in phosphorus after washing, and 59 exhibiting an increase. Again, the contrast between the Mist Mountain and Gates formations is not as important as it first ap-

pears, because of the different analytical procedures referred to above.

The large volume of Gates Formation data also permits some seam-by-seam comparisons. In Figure 11, data from the coal seams from one Gates Formation deposit are arranged in stratigraphic order. For each seam there are three stacked bars, the top one representing the percentage of samples in which phosphorus decreased during washing, the middle one the samples which showed no change, and the bottom bar the samples in which phosphorus increased. In most seams more than 70 per cent of the samples exhibit no change, that is, delta P is between -0.01 and +0.01. However, there are two examples in which more samples show an increase (delta P > +0.01) than a decrease (the fourth and sixth seams up from the base). The seam situated between them, in contrast, has the highest proportion of samples showing a decrease. Even in these relatively extreme examples, however, the largest group of samples showed no change.

DISCUSSION

TRENDS IN PHOSPHORUS CONCENTRATION IN RAW COALS

Raw Mist Mountain Formation coals have higher mean phosphorus concentrations than the Gething Formation coals, which in turn contain more than the Gates Formation. There are no consistent systematic variations in phosphorus concentrations with stratigraphic position in any of the formations, although in the cases of both the Mist Mountain and Gates, seams from the lower half of the formation generally contain, on average, less phosphorus than those from the upper half. Another trend is that the base of the Mist Mountain Formation appears to contain coals relatively low in phosphorus and lacking phosphate minerals. In all coking coal bearing formations raw coal data are highly variable on all scales.

Another interesting result, obtained from the channel sample data, is that variations in phosphorus with stratigraphic position are similar to variations in the ash contents of the samples for both the Mist Mountain and Gates formations. This suggests that any effect of stratigraphic position on phosphorus content can be overridden by the influence of variations in the ash content of individual samples, which has implications for the nature of association of the phosphorus.

FORMS OF ASSOCIATION OF PHOSPHORUS

Phosphorus in coals, especially higher rank coals, is generally believed to be associated predominantly with inorganic material (Burchill *et al.*, 1990), although a small but uncertain proportion may be associated with the organic fraction (Swaine, 1990; Finkelman, 1980). The phosphorus-containing mineral phases in coals include: apatite, most commonly fluorapatite (Swaine, 1977); crandallite-series minerals; monazite; and xenotime (Finkelman, 1980). Several recent studies (*e.g.*, Finkelman, 1980; Lyons *et al.*, 1990) have stressed the relative importance of very small (micron and submicron-sized) accessory mineral grains, including phosphates, dispersed throughout the organic matrix, as important sites for trace elements in coal.

Four types of evidence have been applied in this study to determine the mode of occurrence of phosphorus in British Columbia coking coals: comparison and correlation with ash content, low-temperature ash mineralogy, correlation with fluorine concentrations, and comparison of phosphorus concentrations in clean and raw coals.

The graphs showing phosphorus in coal *versus* ash and phosphorus in ash *versus* ash (Figures 5 to 9), as mentioned earlier, can not be interpreted with certainty. As noted, however, some of the latter type of graph appear to have two distinct components, a line with a high negative slope at low ash contents, and a horizontal line at higher ash contents. This type of relationship has been noted elsewhere (Nicholls, 1968; Finkelman, 1980), and has, in some cases, been interpreted to represent two types of association, organic association in the low-ash range, and inorganic otherwise. As pointed out by Nicholls (1968), it is most appropriate to apply element/ash diagrams to data from one seam and preferably from one location, and the Line Creek ply-by-ply samples qualify. Based on data for 8 and 9-seams (Figures 5f and 5h), it is apparent that the samples in which accessory apatite was identified in greater than trace amounts (large dots), are essentially those low-ash samples which form the segment of the graph with the high negative slope, that is, the portion which would be ascribed to organic association using Nicholls' approach. If all these samples were removed from Figures 5f and 5h, the remaining data would plot on horizontal lines, the relationship indicative of inorganic association.

These examples demonstrate clearly that another critical variable must be considered in interpreting graphs like those in Figures 5 to 9, and that is the mineralogy of the inorganic fraction. As noted by Finkelman (1980), the presence of a very small amount of an accessory mineral containing a trace element, in this case apatite, is capable of producing a significant increase in the concentration of that element, with only a negligible increase in ash content. This effect probably has influence not only in the Line Creek 8 and 9-seam samples, but also in other instances where this type of two-component graph occurs (for example, Figure 8f). This verifies that the main weakness of this approach is that it wrongly assumes that the trace element (phosphorus in this case) concentration in each ash increment remains constant (Nicholls, 1968; Finkelman, 1980).

Data here therefore suggest that phosphorus in British Columbia coking coals is associated with the inorganic fraction, primarily with apatite minerals, including fluorapatite. More support for this conclusion comes from the positive correlations between phosphorus and fluorine (Figure 10), most notably for Mist Mountain Formation samples.

The comparisons between phosphorus contents in raw and clean samples of Mist Mountain coals also tend to confirm an inorganic association. For example, the mean phosphorus concentration in clean coal is substantially lower than the mean in raw coals. Moreover, where phosphorus values are available on both raw and clean coal from the same samples, the majority of clean Mist Mountain coals contain less phosphorus than corresponding raw coals.

In the case of the Gething Formation, it was not possible to compare raw and clean coal from the same samples, but the substantially lower mean phosphorus concentration in clean coals, compared to raw coals, suggests an inorganic association.

The situation in the Gates Formation appears to be different - the overall mean phosphorus concentration in clean coals is essentially the same as that in raw coals, and there is a higher proportion of samples for which removal of phosphorus is not achieved during washing, or for which phosphorus concentrations actually increased during washing. Despite this outcome, there is no valid basis for concluding that the association of phosphorus in Gates coals is fundamentally different. As mentioned previously, the sample preparation techniques most commonly used for the Gates samples would tend to liberate and separate relatively less mineral matter than those used for the coals from the other formations. Moreover, the relationships between phosphorus and ash, and between phosphorus and fluorine, are similar to those derived from the other formations, suggesting that the association of phosphorus in the Gates Formation coals is similar.

It is also important to point out that these results do not imply that the current product coals from mines in northeastern British Columbia contain more phosphorus than the products from the southeast mines. In fact, products from the two regions are not distinguishable in terms of their phosphorus content (Price and Gransden, 1987, Appendix B). The range of phosphorus concentrations in coking coals shipped from the province is covered in the next section.

Even if a direct comparison between the Mist Mountain and Gates clean samples were valid, there are still conditions which could lead to differences in trace element behaviour of the type seen here, without having to invoke a fundamental difference in the manner of association of phosphorus. Finkelman (1980) and Swaine (1990) have summarized the weaknesses in the washability approach to determining an element's affinity in coal, in particular noting that elements associated with mineral grains which are finely dispersed through the organic matrix (macerals) will behave as though they are associated with the organic fraction. Given the important influence of small amounts of accessory mineral grains on trace element concentrations, it would be extremely surprising if trace elements in coals from different geological formations behaved identically in float-sink tests.

The contrasts in the washability behaviour of phosphorus in individual Gates Formation seams (Figure 11), suggest that the influence of the degree of dispersal and grain size of mineral grains noted in the previous paragraph may in fact be operating, with the seams which demonstrate more increases than decreases perhaps containing a greater percentage of their phosphorus in finely dispersed grains.

The crandallite-series mineral gorceixite, which was identified in the low-temperature ash of one Mist Mountain Formation sample, occurs in some tonsteins found in the Kootenay coalfields, as does apatite, though less frequently. This suggests a possible volcanic source for some of the phosphorus in British Columbia coking coals, as the tonsteins are believed to have a volcanic origin (Goodarzi *et al.*, 1990).

COMPARISON WITH WORLD COALS

The range in phosphorus values in metallurgical coal products from British Columbia is 0.023 to 0.079 per cent, with an unweighted average of 0.046 per cent (figures derived from ash analysis data in Appendix B of Price and Gransden, 1987). The calculated means of clean coal phosphorus data in this study (0.030 to 0.044 per cent) are close to this product average, and in the case of the Gates Formation the mean corresponds almost exactly. Overall, a phosphorus concentration on the order of 0.05 per cent is a representative average for our clean coking coals. With raw coals it is not possible to determine a single average, but mean phosphorus contents in the three metallurgical coal bearing formations range from about 0.04 to 0.08 per cent.

The estimated range of phosphorus contents in most world coals is on the order of 0.001 to 0.3 per cent (Swaine, 1990), and an estimated world-wide average is 0.05 per cent (Bertine and Goldberg, 1971). The mean concentrations of phosphorus in British Columbia's coking coals, clean and raw, clearly compare favourably with both this range and average. Moreover, phosphorus contents in British Columbia coals do not appear to be anomalous when compared with ranges of values of coals from Australia, South Africa, Europe, U.S.A., U.K. and other regions (*see* Swaine, 1990, Table 5.23). Phosphorus concentrations in 200 samples of Bowen Basin (Queensland) coal, for example, range from 0.001 to 0.35 per cent.

When comparing the mean concentrations of phosphorus in our coals with mean values from other countries some contrasts are apparent. For example, coals from the eastern United States, including Appalachian coals cited by Finkelman (1980), for which the mean in 754 raw(?) samples is 0.018 per cent, tend to contain less phosphorus than our coals. The average phosphorus content in British

coals is 0.025 per cent (British Coal Corporation, unpublished data, *in* Burchill *et al.*, 1990), while the average phosphorus concentration in coals from New South Wales and Queensland is 0.031 per cent (Swaine, 1977). British Columbia coals thus appear to contain relatively higher mean concentrations of phosphorus, but by factors much less than an order of magnitude, than coals from some other parts of the world.

MARKETING AND PRODUCTION IMPLICATIONS

Whether phosphorus represents a negative factor in the marketing of coking coals depends on many other variables, as noted in the Introduction. The large volume of British Columbia coking coal currently being sold on the world market (in excess of 20 million tonnes per year) suggests it is not in general a significant problem. Specific users may have more stringent specification require-

ments, however, and achieving success in these markets may involve tight production control.

The data summarized here suggest that, where limiting phosphorus in raw coal is a concern, there appears to be some potential in selectively mining seams from particular parts of the stratigraphy. However, careful quality control sampling, even within a single seam, would be necessary to overcome the wide variabilities in phosphorus concentrations.

Phosphorus concentration in a clean coal depends on the concentration in the raw coal, as well as the response of the coal to beneficiation. The good news here is that the phosphorus in Kootenay and Peace River coking coals appears to be mainly inorganically associated, implying that separation during coal processing should, in general, be relatively simple. However, there appear to be inconsistencies in the responses of individual samples or seams to washing, probably related to the variations in ease of liberation of the apatite mineral grains.

CONCLUSIONS

Calculated mean phosphorus concentrations in raw coals of the Mist Mountain, Gething and Gates formations are 0.076 per cent, 0.063 per cent and 0.042 per cent, respectively. There is no consistent stratigraphic trend in mean phosphorus concentrations in the Mist Mountain Formation, but seams in the upper half appear to contain, on average, more phosphorus than those from the lower half, and seams in the basal part of the formation are relatively low in phosphorus. In the Gates Formation also, seams from the upper part of the section tend to contain higher concentrations of phosphorus. In all examples studied, variations in raw phosphorus concentrations within samples from a single seam can be large.

Phosphorus in British Columbia coking coals is predominantly associated with inorganic material, chiefly apatite minerals, including fluorapatite. The two main factors affecting the phosphorus content of a given coal are the amount of ash and the mineralogy of the accessory minerals. A representative average phosphorus content in clean coking coals is 0.05 per cent.

Future work will focus on accurately determining the actual sites that phosphorus occupies within the coal, and this will lead to a better understanding of how to control this element in British Columbia's coking coals.

ACKNOWLEDGMENTS

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APPENDIX

PROXIMATE AND PHOSPHORUS DATA AND ASH MINERALOGY

Sample Number	Property	Pit	Seam Name	Ash %	Mols. %	V.M. %	F.C. %	P %	Mineralogy of Low-Temperature Ash (LTA)
						(A.D. BASIS)			
Q8901	FORDING	EAGLE	15	8.43	1.40	31.88	58.29	0.026	Quartz, kaolinite, illite/muscovite, calcite, fluorapatite, trace: – siderite?
Q8902	FORDING	EAGLE	14-9	21.18	1.12	28.76	48.94	0.118	Quartz, kaolinite, siderite, illite, pyrite
Q8903	FORDING	EAGLE	14-0	6.25	1.36	29.07	63.32	0.162	Quartz, kaolinite, gorceixite?, fluorapatite, siderite, trace: – pyrite, amphibole?
Q8904	FORDING	EAGLE	14-2	14.71	1.13	27.49	56.67	0.079	Quartz, kaolinite, dolomite/ankerite, illite, trace: – siderite, apatite, pyrite
Q8905	FORDING	TAYLOR	7	25.42	0.72	20.00	53.86	0.197	Quartz, kaolinite, siderite, illite, trace: – fluorapatite, pyrite?
Q8906	FORDING	TAYLOR	5	17.64	0.77	19.72	61.87	0.039	Quartz, kaolinite, illite, trace: – siderite, pyrite?
Q8907	FORDING	TAYLOR	9	19.61	1.25	21.21	57.93	0.162	Quartz, kaolinite, siderite, trace: – illite, pyrite, apatite?
Q8908	FORDING	POND	TAILINGS	21.35	1.12	20.12	57.41	0.070	Quartz, kaolinite, illite, magnetite, trace: – siderite, dolomite/ankerite, pyrite
Q8909	BYRON CREEK	12	MAMMOTH	21.71	0.96	21.77	55.56	0.031	Quartz, kaolinite, illite, calcite, dolomite/ankerite, trace: – siderite
Q8910	BYRON CREEK	51	MAMMOTH	21.32	0.87	22.68	55.13	0.035	Quartz, kaolinite, calcite, dolomite/ankerite, illite, trace: – siderite, rhodochrosite?
Q8911	BYRON CREEK	14	MAMMOTH	21.50	1.09	22.17	55.24	0.013	Quartz, kaolinite, dolomite, calcite, illite, trace: – siderite
Q8912	LINE CREEK	MAIN	10A (TOP)	55.97	0.57	13.59	29.87	0.074	Quartz, kaolinite, illite, trace: – pyrite
Q8913	LINE CREEK	MAIN	10A	26.50	0.61	17.58	55.31	0.035	Quartz, kaolinite, illite/mica, trace: – anatase
Q8914	LINE CREEK	MAIN	10A	17.32	0.61	18.08	63.99	0.013	Kaolinite, quartz, trace: – ilmenite?
Q8915	LINE CREEK	MAIN	10A	17.62	0.64	19.13	62.61	0.017	Quartz, kaolinite, trace: – illite, siderite
Q8916	LINE CREEK	MAIN	10A (BASE)	12.36	0.72	19.75	67.17	0.031	Quartz, kaolinite, trace: – siderite
Q8917	LINE CREEK	MAIN	10B (TOP)	15.49	0.82	19.51	64.18	0.009	Quartz, kaolinite, trace: – illite, siderite, calcite
Q8918	LINE CREEK	MAIN	10B	13.99	0.81	19.92	65.28	0.048	Quartz, kaolinite, calcite, trace: – siderite, pyrite, apatite
Q8919	LINE CREEK	MAIN	10B	24.86	0.72	17.28	57.14	0.022	Quartz, kaolinite, illite, trace: – siderite, anatase
Q8920	LINE CREEK	MAIN	10B	10.94	0.72	21.14	67.20	0.009	Quartz, kaolinite, trace: – calcite
Q8921	LINE CREEK	MAIN	10B (BASE)	15.80	1.22	19.79	63.19	0.013	Quartz, kaolinite, illite
Q8922	LINE CREEK	MAIN	9 (TOP)	12.62	0.82	21.12	65.44	0.026	Quartz, kaolinite, trace: – siderite, pyrite
Q8923	LINE CREEK	MAIN	9	18.34	0.88	19.92	60.86	0.039	Quartz, kaolinite, illite, fluorapatite, trace: – pyrite
Q8924	LINE CREEK	MAIN	9	12.88	0.78	19.61	66.73	0.061	Quartz, kaolinite, fluorapatite, trace: – pyrite
Q8925	LINE CREEK	MAIN	9	5.66	0.92	20.90	72.52	0.079	Quartz, kaolinite, fluorapatite, trace: – pyrite?
Q8926	LINE CREEK	MAIN	9	12.41	0.84	21.33	65.42	0.013	Quartz, kaolinite, illite, trace: – siderite
Q8927	LINE CREEK	MAIN	9	25.78	0.70	22.28	51.24	0.039	Quartz, siderite, kaolinite, trace: – illite
Q8928	LINE CREEK	MAIN	9	7.28	0.68	20.22	71.82	0.122	Quartz, kaolinite, fluorapatite, trace: – siderite, pyrite
Q8929	LINE CREEK	MAIN	9 (BASE)	22.20	0.58	18.50	58.72	0.044	Quartz, kaolinite, illite, trace: – anatase?
Q8930	LINE CREEK	MAIN	8 (TOP)	49.02	0.67	14.81	35.50	0.044	Quartz, kaolinite, illite, trace: – anatase?
Q8931	LINE CREEK	MAIN	8	13.07	0.66	20.37	65.90	0.131	Quartz, kaolinite, fluorapatite, trace: – illite, siderite
Q8932	LINE CREEK	MAIN	8	24.64	0.56	19.10	55.70	0.009	Kaolinite, quartz, trace: – illite
Q8933	LINE CREEK	MAIN	8	22.42	0.61	21.40	55.57	0.013	Kaolinite, quartz, trace: – illite
Q8934	LINE CREEK	MAIN	8	27.24	0.56	20.16	52.04	0.022	Kaolinite, quartz, trace: – siderite
Q8935	LINE CREEK	MAIN	8	8.41	0.60	23.73	67.26	0.052	Quartz, kaolinite, trace: – siderite
Q8936	LINE CREEK	MAIN	8	15.37	0.61	19.40	64.62	0.026	Kaolinite, quartz, trace: – siderite, illite
Q8937	LINE CREEK	MAIN	8	11.63	0.69	20.84	66.84	0.017	Quartz, kaolinite, siderite
Q8938	LINE CREEK	MAIN	8	24.81	0.73	19.14	55.32	0.009	Quartz, kaolinite, illite, siderite, trace: – anatase?
Q8939	LINE CREEK	MAIN	8	11.15	0.73	19.96	68.16	0.013	Quartz, kaolinite, siderite, trace: – fluorapatite, anatase?
Q8940	LINE CREEK	MAIN	8	17.34	0.62	20.26	61.78	0.114	Quartz, kaolinite, illite, fluorapatite, trace: – siderite?
Q8941	LINE CREEK	MAIN	8	13.40	0.58	21.88	64.14	0.066	Quartz, kaolinite, trace: – siderite, apatite, pyrite?
Q8942	LINE CREEK	MAIN	8	13.37	0.59	19.66	66.38	0.022	Kaolinite, quartz, trace: – dolomite/ankerite, siderite, anatase?
Q8943	LINE CREEK	MAIN	8	10.88	0.66	19.31	69.15	0.341	Quartz, kaolinite, fluorapatite, trace: – siderite?
Q8944	LINE CREEK	MAIN	8	12.44	0.59	22.62	64.35	0.092	Quartz, kaolinite, fluorapatite, trace: – illite
Q8945	LINE CREEK	MAIN	8	19.77	0.61	19.21	60.41	0.017	Kaolinite, quartz, trace: – illite
Q8946	LINE CREEK	MAIN	8	17.21	0.61	19.76	62.42	0.031	Kaolinite, quartz, trace: – illite, siderite
Q8947	LINE CREEK	MAIN	8	13.07	0.59	25.13	61.21	0.070	Siderite, kaolinite, quartz, trace: – apatite?
Q8948	LINE CREEK	MAIN	8 (BASE)	56.54	0.72	13.73	29.01	0.026	Quartz, kaolinite, illite, trace: – pyrite, anatase?
Q8949	GREENHILLS	BLACKTAIL	25 EAST	7.63	1.49	29.96	60.95	0.035	Quartz, kaolinite, illite, trace: – siderite, pyrite?, apatite?
Q8950	GREENHILLS	BLACKTAIL	16	16.84	0.93	25.71	56.52	0.039	Quartz, kaolinite, illite, trace: – pyrite, apatite
Q8951	GREENHILLS	N COUGAR	22 UPPER	6.12	3.33	28.26	62.29	0.035	Quartz, kaolinite, anatase?, trace: – siderite, apatite?
Q8952	GREENHILLS	BLACKTAIL	20 UPPER	32.72	0.80	22.90	43.58	0.057	Quartz, kaolinite, illite, siderite, trace: – pyrite, apatite, anatase?
Q8953	GREENHILLS	FALCON	1	9.25	0.52	24.79	65.44	0.083	Quartz, kaolinite, siderite, trace: – apatite, pyrite?
Q8954	GREENHILLS	FALCON	3	19.25	0.51	22.92	57.32	0.135	Quartz, kaolinite, siderite, trace: – pyrite, apatite?, illite
Q8955	BALMER	CAMP 8 EX	7R1	23.94	0.43	20.00	55.63	0.170	Quartz, kaolinite, illite, siderite, trace: – apatite, pyrite?
Q8956	BALMER	CAMP 8 EX	7RX	32.06	0.41	18.30	49.23	0.114	Quartz, kaolinite, illite, trace: – siderite, pyrite?
Q8957	BALMER	CAMP 8 EX	7.S	25.96	0.55	21.70	51.79	0.114	Quartz, kaolinite, illite, siderite, trace: – apatite, pyrite
Q8958	BALMER	CAMP 8 EX	8UX	34.74	0.34	18.77	46.15	0.227	Quartz, kaolinite, illite, siderite, trace: – fluorapatite, pyrite, anatase?
Q8959	BALMER	BALDY 3, 4	4	24.83	0.57	19.76	54.84	0.118	Quartz, kaolinite, illite, trace: – fluorapatite, pyrite, siderite?
Q8960	BALMER	ADIT29E	8UC	28.03	0.43	18.47	53.07	0.118	Quartz, kaolinite, illite, siderite, trace: – dolomite?
Q8961	BALMER	POND	TAILINGS	38.28	0.42	18.01	43.29	0.066	Quartz, kaolinite, illite, trace: – dolomite/ankerite, siderite, pyrite?
Q8962	QUINTETTE	WOLVERINE	J3	13.77	0.86	21.24	64.13	0.013	Quartz, kaolinite, siderite, illite, trace: – apatite?
Q8963	QUINTETTE	WOLVERINE	G2	16.31	0.74	21.19	61.76	0.052	Quartz, kaolinite, dolomite/ankerite, trace: – siderite
Q8964	BULLMOOSE		A2	12.51	0.71	24.72	62.06	0.017	Quartz, kaolinite, siderite (manganoan), dolomite/ankerite
Q8965	BULLMOOSE		B	11.05	0.77	24.93	63.25	0.031	Dolomite, quartz, calcite, kaolinite, trace: – pyrite, plagioclase?
Q8966	BULLMOOSE		A1	21.34	0.76	23.63	54.27	0.009	Quartz, kaolinite, illite, dolomite, pyrite
Q8967	BULLMOOSE		C	28.13	1.00	19.90	50.97	0.114	Quartz, illite/muscovite, kaolinite, dolomite/ankerite, trace: – pyrite, magnetite?
Q8968	BULLMOOSE		D	22.14	1.00	23.40	53.46	0.052	Quartz, kaolinite, dolomite, illite, trace: – pyrite, magnetite?
Q8969	BULLMOOSE		E	21.68	1.10	24.42	52.80	0.057	Quartz, kaolinite, illite, trace: – dolomite, pyrite, siderite, apatite?

Note: All results are based on raw coal samples and are not representative of clean, product coals.